### **Acoustic Transduction Materials and Devices**

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#### LONG TERM GOALS

Develop improved, more reliable air and underwater acoustic transducers, encompassing a wide frequency spectrum from 10 Hz to 100 MHz with systematic and conjunct studies on materials, devices, modeling, and electronics.

### **OBJECTIVES**

Develop high performance piezoelectric materials (high strain, agile, low loss piezoelectrics) and related ecological materials with improved reliability for better transduction devices.

The applications are to Cymbal and Tonpilz transducer arrays for 3 - 50 kHz sonars, thin/thick film transducers for 10 - 100 MHz medical acoustic devices, underwater intensity sensors for low-frequency surveillance, and flexural amplification mechanisms of the solid state strain for air acoustic transduction. Modeling and computer simulations of domain motions and structural vibrations and supporting electronics will be developed.

#### **APPROACH**

The Materials Research Laboratory (MRL) at PSU is developing piezoelectric materials and actuator components, including improved lead magnesium niobate (PMN) - lead titanate, and other relaxor electrostrictors for multilayer Tonpiltz transducers, flextensional cymbal transducers, and thin film Lead Zirconate Titanate (PZT)-based ceramics. The reliability issues of ceramic actuators such as high voltage, power and stress dependence are explored in addition to the destruction monitoring system using acoustic emission. Ultrasonic motors are developed for future actuation technology. Modeling and computer simulation studies are conducted in terms of ferroelectric domain structures and structural vibrations. The Applied Research Laboratory (ARL) is developing Cymbal arrayed projectors, PMN Tonpilz tunable transducers, thin/thick film micro-Tonpilz transducers and controlling electronics. The Center for Acoustics and Vibrations (CAV) develops underwater acoustic intensity probes, integrated mechanical amplifiers and frequency-agile flexural transducers. Monthly technical meetings coordinate a unique combination of strengths in transducer design and in transducer materials.

# WORK COMPLETED

#### A. Materials Studies

#### Transducer materials

The phase diagrams of several relaxor-based systems have been explored for use in piezoelectric/electro-strictive transducers: PIN-PMN, PSN-PT, PIN-PT, PIT-PT. The system (1-x)

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Form Approved OMB No. 0704-0188 PIN: x PMN (lead indium niobate: lead magnesium niobate) has been shown to display non-hysteretic strain behavior. The composition x = 0.50 exhibited the strain  $8.12 \times 10^{-4}$  for a 45 kV/cm field, and the resulting electrostriction coefficient, Q33, was  $1.42 \times 10^{-2}$  m<sup>4</sup>/C<sup>2</sup>. Continuing work has also focused on the relaxor:PT (lead titanate) materials for piezoelectric applications. One material showing promising piezoelectric properties has been (1-x)PSN:x PT. At the morphotropic phase boundary (MPB), ceramics have shown strain levels of nearly  $60.0 \times 10^{-4}$  at 90 kV/cm. The piezoelectric properties  $(d_{33}, k_p, \text{ and } k_t)$  have also been found to have attractive values: 595 pC/N, 82%, and 57%, respectively. A related MPB system, (1-x)PIN:x PT, has also been explored. The MPB separating the pseudo-cubic and tetragonal phases was determined to be located at x = 0.38 and showed some temperature dependence. At the MPB the piezoelectric coefficient d<sub>33</sub> and coupling factor k<sub>31</sub> were 395 pC/N and 30%, respectively.

Concerning the tungsten bronze, the crystallographic details including the ionic coordination, the site occupancies and the thermal anisotropy of compositions of a lead containing tetragonal solid solution system (PBN) near the MPB were determined by single crystal x-ray diffraction to understand the polarization mechanisms in the structure that contains the polarization component in off-axis direction, or zigzag polarization.

# Reliability

Since the ceramic actuators are occasionally used under high voltage/power and high stress, systematic data accumulation on these characteristics is important to realize reliable transducer materials. We improved our piezoelectric measuring system using a resonance technique at a constant current (i.e. constant vibration level) condition. During this period, the ceramics of lead magnesium niobate with lead titanate (PMN-PT) near the morphotropic phase boundary have been examined with Mn or Fe doping to test the acceptor doping effect. The properties of those samples under low-level and high-level drive have been compared. When the temperature rise is around 20°C, the vibration velocity around 0.3 m/s can be obtained for a sample with 0.5 wt.% Mn doping.

The effect of mechanical pre-stress on the electromechanical properties of 0.9PMN-0.1PT ceramic over a broad temperature and DC bias field range has been measured. In the investigation, the compressive stress was applied in both parallel and perpendicular to the electric field direction. Also using neutron diffraction and dilatometry, both the intrinsic electrostrictive coefficients (lattice coefficients) and the contribution from the nano-polar regions were determined. The shear electrostrictive coefficient Q44 was determined from the Neutron data. It was shown that the intrinsic volumetric electrostrictive coefficient  $Q_h$  for PMN-PT is on the same level as those of normal ferroelectrics. The measured electrostrictive coefficient  $Q_h$  from the macroscopic strain response, which is much smaller than that of the lattice  $Q_h$ , is a direct consequence of the large contribution of the nanopolar regions to the macroscopic polarization response of the material.

# **B.** Composite Systems

### Cymbal transducers

We studied the design, fabrication, modeling, and calibration of cymbal-type transducers and arrays as underwater electroacoustic projectors and receivers. The cymbal transducer consists of a piezoelectric ceramic (PZT) disk poled in the thickness direction which is mechanically coupled to two thin metal caps on each of its electroded faces. Each cap is shaped such that a shallow air-filled cavity exists

between the cap and the ceramic after they are attached. These caps serve as mechanical transformers for converting and amplifying the small radial displacement and vibration velocity of the piezoceramic disk into a much larger axial displacement and vibration velocity normal to the surface of the caps.

One of the distinct advantages of the cymbal transducer is the ability to readily tailor its resonance frequency simply by changing the cap material or dimensions. An easily "tunable" double resonance behavior can be generated simply by capping the PZT disk asymmetrically. Single element transducers have been evaluated under hydrostatic pressure up to 7 MPa to ascertain both performance capability and means of failure. Cymbals with stiffer caps, thicker caps, and caps with deeper cavities can tolerate higher pressures before failure. Using an underwater anechoic test chamber, the hydrophone and projector performance of these transducers has been evaluated both as single elements as well as in the form of small arrays. Specifically, the effects of different cap materials, cap shape, PZT type, and potting on the electroacoustic performance were examined. The measured and calculated results of the cymbal transducer array show it to be a viable medium to high power transducer for shallow water projector applications from 5 kHz to about 50 kHz.

Another variant of the cymbal design—concave cymbals—is under study. When subjected to high pressures, the standard cymbal fails when the metal caps collapse. This limits the use of cymbal and moonie transducers in the deep ocean. The main feature of the new design is the reversal of the caps from convex to concave orientation while replacing the piezoelectric disk with a piezoelectric ring. This greatly increases the pressure tolerance while maintaining high sensitivity.

# Acoustic intensity probes

The two-geophone underwater acoustic intensity sensor, "u-u probe," has been investigated. This intensity probe concept contains two magnetically shielded, neutrally buoyant geophones that are suspended by elastic members inside a flow- noise suppressing cylindrical rubber shell filled with water. Neutrally buoyant sensors are required so that a true measurement of the acoustic particle velocity is obtained. This is accomplished by casting the geophones in positively buoyant syntactic foam. A thin layer of " $\mu$  metal" is used to shield the two independent magnetic fields of the geophones. The elastic suspension system provides stiffness in the radial direction and virtually no stiffness in the axial direction. All outer shell materials are selected to be acoustically transparent underwater. The signal processing equations were derived for velocity sensitive or acceleration sensitive sensors. Prototypes have been fabricated and evaluated. A standing-wave calibrator for acoustic intensity sensors was designed, developed and tested. It is adequate for calibrations that can be performed in a standing-wave field. Work is completed on the u-u probe; emphasis is shifting to the acoustic vector sensor.

# Film transducers

The goal is to use the tonpilz design to facilitate development of high frequency transducers (30 - 100MHz) without requiring difficult thinning of the bulk ceramic actuator. By utilizing thin/thick films and micro-fabrication techniques, it will ultimately be possible to integrate a transducer array with on-chip amplification electronics. The project is separated into three areas: tonpilz transducer design, micro-tonpilz fabrication, and electronics.

The modified KLM model has been developed and verified and is being used as the primary design model. It includes varying areas from the ceramic to the radiating surface, adding quarter wave matching layers, inductor tuning, and a user defined input pulse. The program outputs are the typical ultrasonic pulse-echo bandwidth and insertion loss, as well as the standard sonar community data

outputs of transmitting voltage response (TVR) and free field voltage sensitivity (FFVS). The area change in the tonpilz design can be thought of as an impedance transformation that is frequency independent. This may allow minimizing the insertion loss with the area change and broadening the bandwidth with a quarter wave matching layer. For comparison, a 2 MHz transducer was modeled using the best design for: 1) solid ceramic, 2) 1-3 composite, and 3) tonpilz. The tonpilz design had a 97% bandwidth and a 0.7 dB insertion loss, which was better than the other materials. An electronic model of the Redwood circuit was developed on PSPICE and verified the match with the KLM model. Two experimental 2 MHz transducers were built using an off-the-shelf 1-3 composites (60% and 33% volume fraction) and head and tail masses using metal shim stock. Theory and experimental pulse-echo responses matched very well giving a 26% bandwidth.

In the area of sol-gel processing, a new method based on a methoxyethanol solvent modified with acetylacetone has been developed. Using a combination of rapid thermal annealing and a post-crystallization heat- treatment step, films with remanent polarizations above  $40\mu\text{C/cm}^2$  and dielectric constants of >1200 at room temperature have been prepared on platinized Si substrates. Films with thicknesses between 0.25 and 7  $\mu\text{m}$  have been characterized. The sol-gel prepared films were found to have little as-deposited polarization, in contrast to sputtered films, which usually have a strong preferred polarization direction. As a result, the sol-gel films showed aging rates (typically ~7 %/decade) which were independent of poling direction, while sputtered films showed a strong asymmetry in aging. There is also growing evidence (based on low temperature dielectric constant data, the dielectric constant measured as a function of the ac drive field, and high field poling) which suggests that as the grain size of the PZT films increases, extrinsic contributions to the dielectric properties become more important.

There was substantial success in improving the wet etching of thick (>1  $\mu$ m) sol-gel deposited PZT for the fabrication of the micro-tonpilz array. While HF-based chemistries etch PZT efficiently, they also leave significant amounts of fluorine-based residue. While HCl-based chemistries leave no such residue, these chemistries excessively attack the PZT grain boundaries even underneath the masking material. We have found that an HF etch followed by a quick HCl cleanup etch at 40°C results in a clean surface with good resolution of the desired PZT structures. Wet etch undercut in the structures appears to be on the order of the thin film thickness. Another challenge in the micro-tonpilz fabrication is the deposition and patterning of the thick (8-25  $\mu$ m) head and tail layers necessary for the devices. For the tail mass, we have explored both electroless- and electroplating of Ni.

The high frequency data acquisition system for transducer evaluation consists of many subsystems which are in various degrees of completion: a high voltage and high speed pulser circuit (60V, 10ns), a time gain compensation circuit and multiplexing and demultiplexing units were designed, fabricated and tested. The algorithms to perform synthetic aperture processing and dynamic apodization processing in various configurations have been developed and tested for functionality.

### C. Device Structures

# PMN Tonpilz transducers

The tonpilz design longitudinal vibrator is used as both a projector and receiver in the 3 - 60 kHz frequency range and will be used to evaluate advanced materials, such as PMN, at frequencies above 10 kHz. A standard tonpilz design with a resonance frequency of 25 - 30 kHz was developed. A PZT-4 element was tested to evaluate calibration data from PMN. Using a DC bias test box, two types of

PMN-PT-La were tested: The first (90/10/1) has its maximum dielectric constant at room temperature; and the second (76/24/1) at 85°C. It is desired to use these electrostrictive relaxor ceramics at the temperature of maximum dielectric constant because this will also be the point of lowest impedance over the operating band and the hence the highest transmit levels. A heat control circuit and winding was developed and attached to the high temperature type 2 PMN. Several preliminary date were obtained under a bias voltage at room temperature and at around 85°C.

# <u>Integrated Mechanical Amplifiers</u>

An optimized design strategy for flextensional devices was developed to design a PZT driven inchworm which, when integrated in a compliant composite material, acts as the driver of a smart material. The inchworm consists of three flextensionals in series that travel between two parallel stationary bars. Extensive testing of the inchworm's performance under different driving signals indicated that while an optimum speed could be obtained over the designed displacement range, the force generated was well below the design expectations. This is attributed to the inability of the clamping elements to oscillate between and clamp-and-release condition at the high frequencies (200 Hz) required.

As a result of shortcomings on the inchworm design (in particular, the low force levels), we redirected our efforts to exploring motion amplification using the locking mechanisms that are commonly used in designing screw devices. The new series of motion amplifiers, called wedgeworms, use PZT stacks as the driving engine and rely on a high friction coefficient (approximately 0.6) to achieve very high driving forces. Our goal is a driving force equal to 1/4 the blocked force of the driving stack, e.g. 1 kN for a 4kN stack blocked force. Our first wedgeworm design is being fabricated.

# Frequency-agile flexural transducers

Frequency agility was pursued in two main projects: an actively-tuned solid-state mechanical vibration absorber and flexural piezoelectric transducers with frequency agility obtained via membrane loads.

We developed a mechanical vibration absorber that actively tunes itself to track a disturbance frequency. The basic principle underlying the operation of the device is the ability to change the mechanical stiffness of a piezoelectric element by changing its electrical boundary conditions. In this case, capacitive shunting is used in order to avoid introducing any unwanted damping, and to maintain high performance. The control system monitors the voltage and current across the piezoelectric element to estimate the disturbance frequency. From this frequency, the controller determines the shunt capacitance needed to tune the natural frequency of the device to the disturbance frequency, then switches it in, using a relay-driven parallel network. Within the tuning range, experiments demonstrated a 20 dB improvement in performance over that of a conventional, non-tunable passive system. In principle, the frequency of a solid-state device could be changed by about 40% from a nominal value.

The bandwidth of a resonant transducer is commonly increased by adding damping to the device, which also decreases its sensitivity. An alternate approach involves active tuning of the resonance frequency over a broad range while maintaining high sensitivity. Compressive in-plane loads were used to alter the natural frequency and coupling coefficient of flexural piezoceramic devices. Two types of transducers were studied, trilaminar beams, and trilaminar disks. Finite element models were developed and used to predict changes in device short and open circuit natural frequencies, as well as coupling coefficients which were also measured. Modifications to the FEM to account for boundary

conditions and stress depoling significantly improved the agreement with experimental results. The inplane loads significantly changed the natural frequencies and coupling coefficients of the transducers. Experiments on the trilaminar piezoceramic beam yielded a 36% change in frequency, and an increase in coupling coefficient of 38%. For the trilaminar axisymmetric piezoceramic disk, experiments yielded a change in frequency of 13%, and an increase in coupling coefficient of 15%. This demonstrated that membrane loads can be used to achieve frequency agility in flexural transducers without loss of sensitivity.

# Ultrasonic motors

Very tiny motors smaller than 1cm are difficult to produce with sufficient energy efficiency. Therefore, a new class of ultrasonic motor using high power ultrasonic energy is gaining wide spread attention. Ultrasonic motors made with piezoceramics whose efficiency is insensitive to size are superior in the mini-motor area. A compact ultrasonic rotory motor as tiny as 3 mm in diameter has been developed, and is a promising candidate for intravascular and intraureteral applications. The stator consists of a piezoelectric ring and two concave/convex metal endcaps with "windmill" shaped slots bonded together, so as to generate a coupled vibration of up-down and tortional type. Since the component count and fabrication process are minimal, the fabrication price should decrease remarkably, and it should be suitable for disposable use. When driven at 160 kHz, a maximum revolution of 600 rpm and a maximum torque of 1 mN·m were obtained for an 11 mm diameter motor.

# **D.** Modeling Studies

# Domain structures

We successfully observed ferroelectric domains using Environmental Scanning Electron Microscopy, which will greatly enhance the ability to study domain microstructures in ferroelectric materials. This technique does not require etching and electroding and could even see the domain structures on a cracked unpolished surface. Using the high resolution CCD microscope, we observed the dynamic domain motions in PZN-PT single crystals with a morphotropic phase boundary composition at various temperatures. We verified the electric field induced phase transition between the rhombohedral and tetragonal phases according to the applied field direction.

### Crystal orientation dependence

We intensively investigated the crystal orientation dependence of electromechanical couplings for various perovskite ferroelectrics both theoretically and experimentally to find the universality of the electromechanical enhancement. Taking a full set of physical parameters of lead zirconate titanate (PZT) systems into account, the crystal orientation dependence of the elastic, dielectric, and piezoelectric constants and electromechanical coupling factors of PZT for various compositions near the morphotropic phase boundary were calculated phenomenologically. For tetragonal PZT, the maximum d33 is always along the spontaneous polarization direction [001]. On the contrary for rhombohedral PZT, the maximum effective d33 is along a direction canted by 50° to 60° away from the spontaneous direction [111]. Near the morphotropic phase boundary on the rhombohedral side, the d33[001]// rapidly increases, while d33[111]// increases only moderately, leading to large enhancement of d33[001]// d33[111]// more than 4. A k33 of more than 90 % can be expected.

The most intriguing result was obtained for another important piezoelectric crystal: single crystal barium titanate (BT). It was found that the effective d<sub>33</sub> was enhanced more than 3.5 times when the electrical field direction was canted by 50° from the spontaneous polarization direction [001]. Along

this direction, the single crystal BT had an effective d<sub>33</sub> as large as 250pC/N, which is comparable with that of PZT. It is noteworthy that this orientation dependence of d<sub>33</sub> and k<sub>33</sub> in BT is very different from the situation in the tetragonal PZT and PT which always have the maximum d<sub>33</sub> and k<sub>33</sub> along the spontaneous polarization direction [001].

# Structural vibration analysis

Three computer models were utilized to predict both the in-air and in-water performance of single element cymbal transducers and arrays, and ultrasonic motor vibrations: (1) ANSYS<sup>®</sup>, a finite element program, to calculate resonance frequencies, structural vibration modes, admittance spectra, and stress distributions, (2) FIRST, a program based on the equivalent electrical circuit of a ring/shell flextensional transducer for obtaining impedance spectra, (3) ATILA, to obtain the dynamic vibration modes for windmill motors.

A comprehensive study on characterization techniques for piezoelectric materials was conducted and a combined measurement technique developed that can accurately characterize all physical properties of piezoelectric materials, including ceramics and single crystal systems in a self-consistent manner.

### **TECHNICAL RESULTS**

The phase diagrams and piezoelectric properties of several relaxor-based systems have been systematically explored for use in piezoelectric/electrostrictive transducers: PIN-PMN, PSN-PT, PIN-PT, PIT-PT.

It was shown from the Neutron data that the intrinsic volumetric electrostrictive coefficient Qh for PMN-PT is on the same level as those of normal ferroelectrics due to the large contribution of the nanopolar regions to the macroscopic polarization response of the material.

The measured and calculated results of the cymbal transducer array showed it to be a viable medium to higher power transducers for shallow water projector applications from 5 kHz to about 50 kHz.

A prototype two-geophone underwater acoustic intensity sensor has been successfully developed.

Using a combination of rapid thermal annealing and a post-crystallization heat- treatment step, films with remanent polarizations above  $40\mu\text{C/cm}^2$  and dielectric constants of >1200 at room temperature have been prepared on platinized Si substrates. We have found that an HF etch followed by a quick HCl cleanup etch at  $40^{\circ}\text{C}$  results in a clean surface with good resolution of the desired PZT structures.

For a flexural fixed-fixed bimorph loaded axially to one-half of its critical buckling load, the resonance frequencies associated with both the voltage and current responses shifted by more than 25%. In addition, the device coupling coefficient increased by 30% at the same load.

Using an optimized design strategy for flextensional devices, an inchworm driven by three PZT stacks has been fabricated.

A semi-actively tuned piezoceramic vibration absorber was developed using a tunable capacitive shunt circuit. Within the tunable frequency band, increases in performance beyond those of a purely passive system were as great as 20 dB and averaged more than 10 dB.

A compact ultrasonic rotory motor ("windmill") as tiny as 3 mm in diameter was developed with a maximum revolution of 600 rpm and a maximum torque of 1 mN·m.

Phenomenological calculation for PZT suggests that near the morphotropic phase boundary on the rhombohedral side, the  $d_{33[001]//}$  rapidly increases, while  $d_{33[111]//}$  increases only moderately, leading to large enhancement of  $d_{33[001]//}$  /  $d_{33[111]//}$  of more than 4. A k<sub>33</sub> of more than 90 % can be expected.

### SYSTEMS APPLICATIONS

High performance piezoelectric materials are directly applicable to sensors and actuators for control of structure borne vibrations. Cymbal and Tonpilz transducer arrays are suitable to diver-held or torpedo guidance imaging sonars and for identifying mines and submarines. Thick film transducers for 10 - 100 MHz are for acoustic imaging components in field-portable medical diagnostic and therapeutic instruments. Acoustic intensity probes and vector sensors are being implemented in various surveillance, platform, and UUV applications. Flexural amplification mechanisms of solid state strain aim for air acoustic transduction applied for anti-radiation and anechoic systems. New inchworm and ultrasonic motors support micro-positioning applications.

#### **TRANSITIONS**

Further study of cymbal arrays is in progress at NRL through ARL/PSU. The underwater intensity probes are now being manufactured by a start-up company (Acoustech) under the SBIR Program. Their use in the NVMS of submarines has been proposed for FY99. A working relation with Blatek, an ultrasonic transducer company, has been initiated so that the micro-tonpilz technology can be commercially demonstrated at the 2-5 MHz frequency range typically used in medical ultrasonics.

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